



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICATIONS : KANNO, et al.
SERIAL NO. : 09/923,373
FILED : August 8, 2001.
FOR : PROCESS FOR PRODUCING COKE, ARTIFICIAL GRAPHITE AND
CARBON MATERIAL FOR NEGATIVE ELECTRODE OF
NON-AQUEOUS SOLVENT TYPE SECONDARY BATTERY AND
PITCH COMPOSITION USED THEREFOR
ART UNIT : 1764
EXAMINER : Tam M. Nguyen

DECLARATION UNDER 37 CFR 1.132

Honorable Commissioner of Patents and Trademarks
Washington, D.C. 20231

Sir:

I, KOICHI KANNO of 22, Wadai, Tsukuba-shi, Ibaraki, Japan, declare that;

1. I graduated from Department of Engineering Science, Faculty of Polymer Chemistry, Master Degree Course of Kyoto University in March 1990 and entered Mitsubishi Gas Chemical Company, Inc. in April 1990. I have been in charge of research and development of a lithium secondary battery in Corporate Research Laboratory from January 1994, and have held the position up to the present time.
2. I am one of the named inventors of the present U. S. Patent Application as identified above, and familiar with the subject matter disclosed in the Application.
3. Experiment

Object of Experiment

The following experiments were carried out, in order to confirm the effect of the addition of sulfur to a negative electrode in the performance of the non-aqueous solvent type secondary battery using a negative electrode comprising a mesophase pitch which is produced by polymerizing a condensed polycyclic hydrocarbon in the presence of HF-BF₃

catalyst.

Procedures of Experiment

In EXAMPLE 2 of the present invention, the performance of the non-aqueous solvent type secondary battery was confirmed by using a negative electrode containing 5 parts by weight of sulfur to 100 parts by weight of the coal tar pitch and 100 parts by weight of the mesophase pitch which is produced by polymerizing a condensed polycyclic hydrocarbon in the presence of HF-BF_3 catalyst. In EXAMPLE 1 of the present invention, the carbon material using the same coal tar pitch and the same mesophase pitch as EXAMPLE 2 of the present invention without containing sulfur was produced. Then the effect of the addition of sulfur to a negative electrode in the performance of the non-aqueous solvent type secondary battery would be confirmed by using a negative electrode comprising the carbon material of EXAMPLE 1.

Experiment

Ninety parts by weight of the graphite powder produced in EXAMPLE 1 of the present invention and 10 parts by weight of a polyvinylidene fluoride powder as a binder were mixed together with dimethylformamide as a solvent. The resultant mixture was applied onto a copper foil, and then dried. The coated foil was cut into 1 cm square to prepare a test specimen for the evaluation. Then, the test specimen was used as an electrode together with an electrolyte solution prepared by dissolving LiPF_6 in a mixed solvent containing ethylene carbonate and diethylcarbonate at a mixing ratio of 1:1 (concentration: 1.0 mol/l), and a separator formed of a 50 μm -thick polypropylene microporous membrane in order to prepare a half cell. Further, in the cell, both a counter electrode and a reference electrode were formed of metallic lithium having a diameter of 16 mm and a thickness of 0.5 mm. The thus prepared half cell was subjected to constant current charge at a current density of 0.2 mA/cm^2 until the electrode potential of the test specimen relative to the reference electrode reached 10 mV. Then, the cell was subjected to constant current discharge at a current density of 0.2 mA/cm^2 until the electrode potential of the test specimen relative to the reference electrode reached 1.5V.

Results of the Experiment;

As the result, it was confirmed that the charge capacity of the cell at initial cycle was 500 mAh/g; the discharge capacity thereof was 300 mAh/g; and the charge-discharge efficiency thereof was 60.0%.

Consideration:

In EXAMPLE 2 of the present invention, the charge capacity of the cell at initial cycle was 347 mAh/g; the discharge capacity thereof was 330 mAh/g; and the charge-discharge efficiency thereof was 95.0%. The carbon material using the same coal tar pitch and the same mesophase pitch as the above experiment containing sulfur is used for producing the electrode in this EXAMPLE 2. Then it is confirmed that a negative electrode of non-aqueous solvent type secondary battery having a high discharge capacity and a high charge-discharge efficiency is produced by using such a sulfur-containing pitch composition.

The addition of sulfur to asphalt is known as described in Hayner, et al (US Patent 6,153,004), however, in Hayner, et al, slurry of solid sulfur in oil, or oil containing sulfur compounds is added to asphalt cement for road pavement in order to avoid the formation of explosive clouds of dust. The present invention is not suggested from Hayner, et al because the technical field and the effect of the addition of sulfur to the carbon material are quite different from Hayner, et al. By the addition of sulfur, the crystallite size L_c was decreased, however, the discharge capacity and the charge-discharge efficiency are increased. Then non-aqueous solvent type secondary battery of high charge-discharge efficiency is produced. This invention has novelty and inventive step because these results are unpredictable.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and beliefs are to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.



Koichi Kanno

Date : June 8, 2005